

Manure Biogas Using Activated Coconut Charcoal Adsorbent and Iron Oxide (Fe_2O_3) Against Carbondioxide (C_2O) and Methane Gas (CH_4)

Yasri Rahmawati¹, Lilik Eka Radiati², Mochammad Junus³

^{1,3}Animal Production, Faculty of Animal Husbandry, Brawijaya University, Malang, East Java, Indonesia-65145

²Animal Products Technology, Faculty of Animal Husbandry, Brawijaya University, Malang, East Java, Indonesia-65145

Email address: yasri1431@student.ub.ac.id, yasriahmawati1431@gmail.com

Abstract — Biogas is an alternative energy source that can be used as heat producing energy. One of the problems contained in biogas is carbon dioxide which can reduce the calorific value of burning biogas, so a purification process is needed. This research aims to determine the levels of methane (CH_4), carbon dioxide (CO_2), N_2 heating value and energy potential in biogas. The results of the paired T-test analysis for the use of adsorbants were not significantly different ($P > 0.05$). Results reduction of methane gas (CH_4), carbon dioxide (CO_2), N_2 from testing cow biogas and chicken manur before and after purification respectively are: the CH_4 content of cow biogas is 19.14% to 46.03%, CH_4 levels chicken manur is 12.07% to 86.55%, N_2 content cow biogas is 0.01% to 0.00%, chicken manure N_2 content is 0.03 % to 0.05%. The calorific value of biogas, the potential biogas energy produced is: the calorific value of cow biogas is 6,282 kJ/L to 15,103 kJ/L, the potential is 247,197 kJ to 594,486 kJ, and the calorific value of chicken manure namely 3,959 kJ/L to 28,399 kJ/L, with a potential of 143,171 kJ to 1,030,907 kJ. It was concluded that the use of coconut shell charcoal and ferrihydrate was effective as a mixed adsorbent in biogas purification.

Keywords— Biogas, carbon dioxide (CO_2), methane gas (CH_4), nitrogen (N_2), purification

I. INTRODUCTION

Utilizing livestock manure as biogas is a way to overcome the energy crisis. Biogas is a renewable energy that can be used as an alternative to reduce the use of fossil energy. Its environmentally friendly nature is the basis for the development of biogas in several countries. Biogas renewable energy can be produced on a large scale in Indonesia because of the abundant potential of raw materials, especially in the livestock sector from chicken and cow manure because it is still not utilized optimally. Livestock manure that can be used to make biogas comes from cattle, chickens, goats, pigs and horses. However, the most frequently used material for making biogas is cow manure.

One of the impacts faced by livestock waste today is the presence of greenhouse gases (GHG), namely methane (CH_4), carbon dioxide (CO_2) and nitrogen (N_2). According to Syarifuddin (2019), the problem of GHGs can contribute to global warming. Biogas produced from the fermentation process of organic waste does not contain 100% combustible gas. Biogas products consist of methane (CH_4) 55-75 %, carbon dioxide (CO_2) 25-45 %, nitrogen (N_2) 0-0.3 %, hydrogen (H_2) 1-5 %, hydrogen sulfide (H_2S) 0-3 %, oxygen (O_2) 0.1-0.5 %, and water vapor.

Of all these elements that play a role in determining the quality of biogas, namely methane gas (CH_4) and carbon dioxide (CO_2) (Ritonga and Masrukhill, 2017). So the biogas content used in the combustion process is methane (CH_4) which has a high calorific value as fuel. The biogas content is very important and can interfere with the combustion process which can reduce the calorific value of carbon dioxide (CO_2) (Samlawi and Sajali., 2021).

Low methane content has low flame quality, only can used as fuel in cooking activities. To increase benefits Biogas as renewable energy requires easy and cheap methane purification stages. According to Niesner et al. (2013) in Fahriansyah et al. (2019), that there are various purification methods developed in the industrial practice of CO_2 separation, namely absorption (physical - purisol, selexol, rectisol, water scrubber; chemical - MEA, DEA, MDEA solvents), adsorption (swing pressure adsorption, TSA), permeation (high pressure and low pressure membranes) and others (cryogenic and biological approaches). Adsorption is a process that occurs when a liquid or gas is bound to a solid and eventually forms a film layer (thin film) on the surface of the solid. Adsorption processes at the surface of a solid involve the transfer of dissolved substances in a gas to a solid surface, where the transfer process is driven by Van der wall forces. According to Yenetekeakis and Goula (2017), the adsorbent that is typically utilized is granular and has a high surface area per unit volume. So, in this research biogas purification was carried out using the adsorption technique, because it has good absorption capacity for the biogas purification process, if technically and economically considered adsorption purification is easy to do. With a methane purification system/equipment, biogas can be applied as a raw material source energy to be converted into electrical energy, heat energy using a co-generator so that it can be used to substitute fuel oil (BBM), electricity which is increasingly expensive.

Solid adsorbents that have the potential to purify biogas are iron oxide (Fe_2O_3) (Fahriansyah et al., 2019), and activated coconut charcoal (Ritonga and Masrukhill, 2017). The application of biogas through direct combustion can speed up the cooking process, because the heat produced is higher and can reduce unpleasant odors and reduce corrosion on engine components. Using purified biogas as fuel can reduce greenhouse gas emissions because burning biogas releases less

nitrogen oxides, hydrocarbons and carbon monoxide than burning gasoline and diesel.

In the research of Ritonga and Masrukhlil (2017), the use of activated charcoal and zeolite adsorbents with various compositions was able to increase the methane content in biogas, while in the research of Guha, et al. (2021) the use of iron oxide (Fe₂O₃) has the potential to be used as an adsorbent to absorb CO₂ well. In this research, the biogas purification method was carried out using a three-layer adsorption technique, because it has good absorption capacity for the biogas purification process.

II. MATERIALS AND METHODS

A. Research Implementation

This research was carried out in August-October 2024 at the Animal Production Laboratory, Brawijaya University. Then the samples were tested at the Agricultural Environmental Instrument Standards Testing Center. The method used in this research is experimental research using a self-assembled biogas purification device. The analysis method uses graphic and descriptive comparisons to describe the relationship between biogas levels before and after purification. The biogas used in this research was chicken and cow biogas taken from laying hen and dairy cow breeders in the Bantur area, Malang Regency. The materials used as adsorbents are activated coconut shell charcoal, ferrihydrate, and foam (silica gel). The tools used are digital scales, cow biogas storage canisters, chicken biogas purification equipment, and an electric motor compressor for transferring cow biogas into canisters.

This study employed an experimental design in which samples were collected both before and after purification.

B. Biogas Formation Process

The anaerobic fermentation process to form bio gas has several phases, namely the hydrolysis, acidification and methanogenesis phases. Attached in Figure 1 is the phase of bio gas formation (Soeprijanto, et al., 2017).

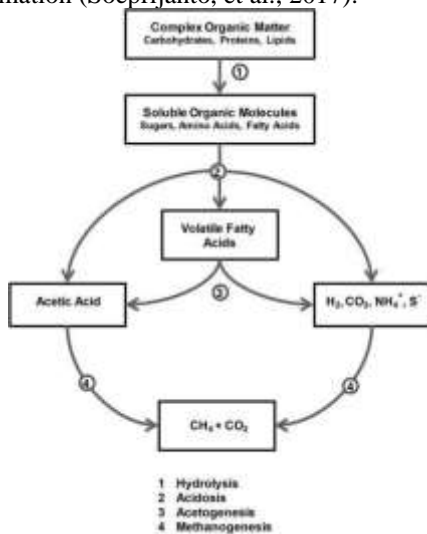


Figure 1 . Biogas Formation Stage

In the hydrolysis stage, organic materials containing hemicellulose, cellulose and other extractive materials such as proteins, carbohydrates and lipids will be broken down into

simpler compounds. To be easily decomposed, complex organic molecules in solid form must first be cut into pieces to facilitate their transportation across the bacterial cell membrane. The results of the hydrolysis phase are simple molecules such as carbohydrates (simple sugars), amino acids and fatty acids. In the hydrolysis phase, the optimal pH ranges from 6 to 7. Apart from that, in the hydrolysis stage the microorganisms that play a role are hydrolytic bacteria and extracellular enzymes such as amylase, lipase, protease and cellulose (Pujiati, et al., 2020).

Before entering the acidification phase, there is a process of breaking down chemical substances such as carbohydrates by enzymes, bacteria, yeast or mold under anaerobic conditions. This is called the acidogenesis phase. The results of the acidogenesis phase are acetic acid (CH₃ COOH), hydrogen (H₂), and carbon dioxide (CO₂) (Soeprijanto, et al., 2017). In the acidification phase, bacteria produce acid which functions to convert short compounds resulting from hydrolysis into acetic acid (CH₃COOH), hydrogen (H₂) and carbon dioxide (CO₂). This bacteria is an anaerobic bacteria that can grow in an acidic atmosphere, namely a pH between 5.5 and 6.5 and works best at a temperature of 30°C. To produce acetic acid, these bacteria need oxygen (O₂) and carbon. For uniform metabolism, a good mixture with a water concentration above 60% is required. Apart from that, these bacteria also convert low molecular weight compounds into alcohol, organic acids, amino acids, carbon dioxide (CO₂), hydrogen sulfide (H₂S) and small amounts of methane (CH₄) (Pujiati, et al., 2020).

The final phase is the formation phase of methane gas (CH₄) or known as methanogenesis. At this point, methane is formed from acetic acid or a compound of hydrogen and carbon dioxide by bacteria. The bacteria that play a role are methanogenic bacteria (methane bacteria). Methanogenic bacteria are anaerobic bacteria whose growth is slower than bacteria in the hydrolysis and acidification phases. The group of methane bacteria includes *Methanobacterium*, *Methanobacillus*, *Methanosarcina*, and *Methanococcus*. Methane bacteria require a digestive tank with a closed and dark atmosphere. Methanogenic bacteria can work well at temperatures of 35°C and are very sensitive to temperature changes of around 2-3°C. The methane formation phase occurs at an optimal pH between 6.5 and 7.5. The end products of metabolism are methane (CH₄) and carbon dioxide (CO₂) from acetic acid, hydrogen gas (H₂) and carbon dioxide (CO₂) which are produced in the acidification phase (Pujiati, et al., 2020). Biogas is generally considered to be low quality natural gas. The methane (CH₄) content of natural gas ranges from 90 to 95% (Yuzheng, et al., 2021). According to Wahyuni, et.al (2009) in Sari, et al., (2020) that the potential of some livestock manure can produce waste according to Table 1.

C. Research procedure

The research procedures carried out were in accordance with research conducted by Suprianti, et al., (2022). The procedure is in accordance with the flow diagram in Figure 2.

TABLE 1. Production of Various Animal Wastes

Types of Livestock	Livestock Weight (kg/head)	Waste Production
Dairy cows	500 – 600	30 – 50
Beef cattle	400 – 500	20 – 29
Broiler	1,0 – 1,5	0,06
Laying Hens	1,5 – 2,0	0,10
Pig	80 – 90	7
Sheep	30 – 40	2

Source: Sari, et al., 2020



Figure 2. Research Flow Diagram

1. Raw Material Preparation

Preparation of processed biogas raw materials. Biogas is obtained from a biogas digester owned by cattle farmers, then stored in polyethylene (PE) plastic. Biogas collection is carried out in the morning, at 10.00 WIB, so that the composition of the biogas will not differ much.

Adsorbent preparation. The activated carbon is first crushed, then aerated to obtain uniform particles of 32 mesh. Then the activated carbon is reactivated under sunlight at daytime temperatures (10.00-13.00) for 2-3 hours, to ensure that no more gas is trapped in the activated carbon. Next, it is cooled and mixed with ferrihydrate adsorbent (Fe₂O₃) then the mixture is put into the adsorption column.

This preparation process is carried out repeatedly according to the variation of the mixture to be used. The total mass of the adsorbent is 2 kg with the variations used as follows.

- Chicken manure biogas samples and cow biogas samples without adsorbents
- Chicken manure biogas samples and cow biogas samples with adsorbent (50% activated charcoal + 50% ferrihydrate (Fe₂O₃)).

2. Test Equipment Preparation

In this research, the biogas purification method was carried out using a three-layer adsorption technique. Foam is placed on top to absorb the moisture from the biogas. The second layer is iron powder (Fe₂O₃) which has paramagnetic properties and functions to absorb CO₂ gas. The final layer is coconut charcoal which has been activated to increase CH₄ in biogas. Gas analysis is carried out after leaving the digester before going through purification and after going through purification. According to Suprianti, et al. (2022), the adsorption column has a capacity of 2,43 liters, with PVC material, a diameter of 2 inches and a column height of 55 cm (40 cm containing the adsorbent mixture). The column is not filled completely with adsorbent, but there is free space at the top so that the adsorption gas is collected before leaving the column. The empty space at the bottom of the column is partitioned with an area filled with adsorbent. The partition is made of wire mesh, which also functions as a support.

The gas purification device assembled to take chicken

biogas samples is shown by (Figure 3), while the cow biogas purification device is shown by (Figure 4). This cow biogas purification tool is equipped with a compressor and biogas storage tube.



Figure 3. Components of chicken manure biogas purification equipmet three absorption layers



Figure 4. Bovine biogas purification cylinders and compressors

3. Adsorption Process

The adsorption process is carried out on a lit stove. The aim is to ensure that the gas that comes out is rich in CH₄, which can be seen from the color of the flame. Biogas that has been stored in the gas container is channeled to the purification device by passing through a rotameter at a rate of 3 liters/minute (Suprianti, et al. 2021).

4. Sampling

Sampling was carried out on two sides, namely on the upstream side before entering the adsorption column and on the downstream side after leaving the adsorption column. Gas sampling was carried out at the 9th minute, because the CH₄ value was greatest at the 9th minute. Samples were taken twice as a backup if the first sample did not show results during testing.

5. Gas Content Test

Gas content was tested using Gas Chromatography at the Agricultural Environmental Instrument Standards Testing Center, Central Java.

D. Research variable

Control variables are methane gas (CH₄), carbon dioxide gas (CO₂), nitrogen gas (N₂), adsorbance effectiveness and heating value.

E. Data Analysis

The data obtained was then analyzed using a paired T-test to assess the effectiveness of using adsorbents. The data obtained is then described to describe the biogas content.

According to Suprianti, et al., (2021), determining performance in the adsorption process can be done by paying attention to the following things:

1. Calorific value of biogas

The calorific value of biogas depends on the CH₄ content in the biogas. Calculation of calorific value using the following formula:

- a. density value of CH₄ is 0.656 kg/m³
- b. The calorific value or LHV (Low Heating Value) of CH₄ is 50,02 MJ/kg (Moran, et al., 2014)
 - Calorific value of CH₄ cow biogas
 - = $LHV \times p \text{ CH}_4$
 - = 32,813.12 Kcal/ltr or 32.813 kJ/m³
 - Calorific value of CH₄ chicken manur
 - = 30262.96 Kcal/ltr or 30.262 kJ/m³ (Anoi, et al., 2022)
- c. Biogas heating value = CH₄ heating value x % CH₄ in biogas
- d. Mass of biogas = v x t x p CH₄ x %CH₄

Information:

- v = gas flow velocity
- p = density (kg/m³)
- t = adsorption time

e. Effectiveness of biogas adsorption

Percentage of carbon dioxide (CO₂)

content resulting from biogas adsorption:

$$= \frac{\% \text{CO}_2 \text{ adsorption input} - \% \text{CO}_2 \text{ adsorption output}}{\% \text{CO}_2 \text{ input adsorption}} \times 100\%$$

III. RESULTS AND DISCUSSION

Testing Methane content was carried out to determine the effect of variations in the adsorbent used on changes in methane gas content in chicken manure and cow biogas which is presented in table 2. The results of the analysis of variance showed that the use of adsorbents in chicken manure and cow biogas did not differ (P>0.05) in relation to CH₄, CO₂, and N₂. The results of gas testing carried out using gas chromatography show that by integrating biogas with adsorbents it will increase CH₄, reduce CO₂ and N₂. The gas chromatography test results can be seen in table 2 and figure 5.

TABLE 2. Ingredients in chicken manure and cow gasbio

Types of Biogas	CH ₄ (ppm)	CO ₂ (ppm)	N ₂ (ppb)
Chicken Manure (A)	399,87	1.237,91	369,14
Chicken manure (B)	158.922,64	10.492,92	389,52
Cow biogas (A)	1.924,25	3.452,33	376,33
Cow biogas (B)	71.890,58	35.809,53	420,62

Information: - Testing Center laboratory test results

Standard Agricultural Environmental Instruments (2023)

- Paired T-test results are not significantly different (P>0.05)
- Sample without adsorbent (A)
- Sample with adsorbent Fe₂O₃ (50 : 50) (B)



Figure 5. Contents of chicken manure and cow biogas

A. CH₄ content there is Livestock Biogas

Figure 5 and table 2 show that the methane gas content in cow biogas before purification is 19,14% or 1.924,25 ppm, while chicken manure before purification is 12,07% or 399,87 ppm, the CH₄ content in this biogas cannot be used as a heat energy, because biogas can burn if the methane gas content is 50-70%. The methane gas content of cows increases after being purified 46.03% or 71,890.58 ppm, while in chicken manure it is 86.55% or 158,922.64 ppm. Gas Cow methane after purification increased by 26.89% and in chicken manure the methane gas content increased by 74.48%. The increase in CH₄ content was caused by a decrease in CO₂ gas contained in biogas (Figure 5). Biogas purification using the absorption method using a mixture of activated charcoal and oxidized iron powder can increase methane gas and reduce CO₂ levels and N₂ contained in biogas after purification.

The composition of methane (CH₄) contained in biogas determines the quantity and quality of the combustion produced. The CH₄ content is generally 40-50% and can be used as fuel for cooking (Sunaryo, 2014), However, the biogas produced contains <40% methane gas, so it needs to be purified. According to Pujiati, et al. (2020) the energy content of biogas depends on the concentration of methane (CH₄). The greater the methane content, the greater the calorific value. Conversely, the smaller the methane content, the smaller the heat contained in it. The biogas content is also influenced by the length of anaerobic fermentation with the help of bacteria such as methanogenic bacteria (Pujiati, et al., 2020). According to Anggito (2014), if methanogenic bacteria experience a lack of nutrition, this can cause inhibition of bacterial growth and reduced methane production.

B. CO₂ Content in biogas and effectiveness

Figure 5 and table 2 show a decrease in CO₂ in chicken manure and cow biogas after purification using charcoal and ferrihydrate adsorbents, this is proven by the carbon dioxide content in cow biogas from 80.86% or 3.452,33 ppm to 53,97% or 35.809,53 ppm, while in chicken biogas the initial carbon dioxide level was 87,93% or 1.237,91 ppm, dropping to 13.45% or 1.0492,92 ppm. Biogas purification using the absorption method using activated charcoal adsorbent can reduce CO₂ levels in cows it was 26,89% and in chickens CO₂ levels fell by 74,48%. This is comparable to research conducted by Samlawi and Sajali (2021) that the use of various types of charcoal in

biogas purification in the form of charcoal media is able to absorb carbon dioxide in biogas which can be used as a medium for biogas purification.

Cow biogas and chicken manure using charcoal and ferrihydrate is 33.25% and 84.70% respectively. This percentage shows the effectiveness of carbon dioxide absorption in biogas after it is purified. Biogas purification uses a mixture of adsorbents in the form of charcoal and ferrihydrate which has a higher absorption capacity for carbon dioxide. If we look at the changes in input and output composition, it can be seen that the use of coconut shell charcoal and ferrihydrate adsorbents (50:50), although not significantly different, has an influence on the effectiveness of CO₂ absorption. This is thought to be because the use of active adsorbants can absorb CO₂. According to Suprianti, et al., (2021) that the greater the surface area and pores of the adsorbent, the greater the adsorption power. Iriani, et al., (2016) explained that the effectiveness of adsorption will be directly proportional to the difference in CO₂ content before and after purification. An increase in pressure will increase the CH₄ composition in the sweet gas and increase the amount of CO₂ absorption.

C. N₂ content in livestock biogas

Figure 5 and table 2 show that the nitrogen (N₂) content in cow biogas before purification is 0.01 % or 376.33 ppb, while chicken manure before purification is 12.07% or

369.14 ppb, the N₂ content in this biogas relatively low, because in general the N₂ content in biogas is 0.3-3 % (Suprianti, et al., 2021), some also say 2.23% (anoi, et al., 2022). The N₂ content of cows decreased after being purified to 0,001 % or 420,62 ppb, while in chicken manure it became 0,05 % or 389,52 ppb. Nitrogen after purification decreased even to almost 0%.

D. Biogas Calorific Value and Biogas Energy Potential

CH₄ content value in biogas is the main thing that determines the calorific value of biogas. The amount of CH₄ content determines the calorific value of biogas. Iriani, et al., (2016) explained that the calorific value will be directly proportional to the methane content in biogas. The results of analysis of variance showed that the use of adsorbents in chicken manure and cow biogas did not differ (P>0.05) in the calorific value and energy potential of biogas. The calorific value and energy potential of biogas are presented in Table 3.

TABLE 3. Calorific value and energy potential of biogas

Types of Biogas	Biogas Calorific Value (kJ/L)	Biogas Energy Potential (kJ)
Cow (A)	6,28 2	247,197
Cow (B)	15,103	594,486
Chicken (A)	3,959	143,171
Chicken (B)	28,399	1.030,907

Note: - Paired T-test results are not significantly different (P>0.05)

- Sample without adsorbent (A)

-Sample with adsorbent Fe₂O₃ (50 : 50) (B)

In Table 3, the calculation results show that the calorific value of cow biogas has increased from 6 , 282 kJ /L to 15,103 kJ/L and chicken manure increased from 3,959 kJ/L to 28,399 kJ/L. According to research by Wiratmana et al., (2012), the experimental and theoretical heating value of biogas is

55,017kJ and 75,034 kJ with a CH₄ composition of 80%. Meanwhile, Suprianti, et al., (2021) produced biogas purified using a mixture of activated charcoal and zeolite (70:30) as an adsorbent of 28.73 MJ/m³ with a biogas energy potential of 775,74 kJ. This is comparable to Iriani, et al., (2016) that purification of 32 mesh size activated carbon produces a heating value of 28,968 kJ/L with a biogas energy potential of 476,524 kJ for 15 minutes equivalent to 0,530 kW-530 Watts. Kusairi and Yangsen, (2015) stated that the electrical power produced by a generator with fuel that has gone through refining is greater than the electrical power produced by a generator with fuel that has not gone through digester refining.

The energy potential of chicken manure biogas produced has increased from 143,171 kJ to 1.030,907 kJ, while for cows it was previously 247,197 kJ to 594,487 kJ. The increase in the calorific value and energy potential of biogas after purification proves that the use of activated charcoal and oxidized iron can increase the heat energy produced. This is comparable to Hariastuti et al. (2014) The level of potassium hydroxide (KOH) compounds in coconut shell charcoal influences the calorific value of biogas, where the higher the level of KOH compounds used, the adsorption ability of activated coconut shell charcoal increases, resulting in a higher biogas calorific value. According to Siregar, et al., as an energy source, bio gas can be burned with a high calorific value, namely in the range of 4700-5000 kcal/m³. According to Apriandi, et al., (2023) said that the calorific value of bio gas is a common indicator used as a benchmark for the quality of bio gas. The calorific value of bio gas depends on how much methane gas (CH₄) is contained in the bio gas. Adiani et al. (2020) state that the calorific value (or energy content) of biogas increases with increasing methane concentration and decreases with decreasing methane concentration. Iriani, et al., (2016) stated that the energy potential of biogas will be influenced by the volume of biogas and the calorific value of biogas.

IV. CONCLUSION

In this study, the use of activated charcoal and ferrihydrate adsorbants in a 50:50 ratio for the purification of cow biogas and chicken manure did not have a significant effect (P>0,05). However, the use of adsorbants can increase CH₄ , reduce CO₂ and N₂ , and increase the calorific value and energy potential of biogas. The content of CH₄, CO₂, O₂, calorific value and energy potential of biogas after purification are:

- a. Cow biogas content, namely: CH₄ (46,03%), CO₂ (53,97%), N₂ (0.00%), calorific value (15,103 kJ/L), and biogas energy potential (594,486 kJ) .
- b. The content of chicken manure is: CH₄ (86,55%), CO₂ (13.45%), N₂ (0,05%), calorific value (28,399 kJ/L), and biogas energy potential (1.030,907 kJ).

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